Bioresource Technology, Vol. 245, Part A, December 2017, pp. 581-589
DOI:10.1016/j.biortech.2017.08.071

Accepted Manuscript

Performance and stability of sewage sludge digestion under CO_2 enrichment: a pilot study.

Luca Alibardi, Kevin Green, Lorenzo Favaro, Peter Vale, Ana Soares, Elise Cartmell, Yadira Bajón Fernández

PII: S0960-8524(17)31376-7
DOI: http://dx.doi.org/10.1016/j.biortech.2017.08.071
Reference: BITE 18680

To appear in: *Bioresource Technology*

Received Date: 18 June 2017
Revised Date: 10 August 2017
Accepted Date: 13 August 2017

Please cite this article as: Alibardi, L., Green, K., Favaro, L., Vale, P., Soares, A., Cartmell, E., Fernández, Y.B., Performance and stability of sewage sludge digestion under CO_2 enrichment: a pilot study., *Bioresource Technology* (2017), doi: http://dx.doi.org/10.1016/j.biortech.2017.08.071

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

Published by Elsevier. This is the Author Accepted Manuscript issued with:
Creative Commons Attribution Non-Commercial No Derivatives License (CC:BY:NC:ND 4.0).
The final published version (version of record) is available online at DOI:10.1016/j.biortech.2017.08.071
Please refer to any applicable publisher terms of use.

ACCEPTED MANUSCRIPT

Title:

Performance and stability of sewage sludge digestion under CO_2 enrichment: a pilot study.

Authors:

Luca Alibardi[a], Kevin Green[a], Lorenzo Favaro[b], Peter Vale[c], Ana Soares[a], Elise Cartmell[a,1], Yadira Bajón Fernández[a*]

Affiliations:

[a] Cranfield Water Science Institute, School of Water, Environment and Energy, Cranfield University, Cranfield, Bedfordshire, MK43 0AL, UK

[b] Department of Agronomy Food Natural resources Animals and Environment, University of Padova, Viale dell'Università 16, 35020, Legnaro, Padova, Italy

[c] Severn Trent Water, 2 St John's Street, Coventry, CV1 2LZ, UK

[*] **Corresponding author:**

Dr. Yadira Bajón Fernández

Cranfield Water Science Institute, Cranfield University, Cranfield, MK43 0AL, UK

E-mail: y.bajonfernandez@cranfield.ac.uk

[1] *Present address*: Scottish Water, Castle House, 6 Castle Drive, Carnegie Campus, Dunfermline, KY11 8GG, UK

ACCEPTED MANUSCRIPT

Abstract

Carbon dioxide (CO_2) injection in anaerobic digestion has recently been proposed as an interesting possibility to boost methane (CH_4) recovery from sludge and organic waste by converting a greenhouse gas into a renewable resource. This research assessed the effects of exogenous CO_2 injection on performance and process stability of single-phase continuous anaerobic digesters. Two pilot scale reactors treating sewage sludge were operated for 130 days. One reactor was periodically injected with CO_2 while the other acted as control. Two injection frequencies and injection devices were tested. The results indicated that CO_2 enrichment allowed an increase in CH_4 production of *ca.* 12%, with a CH_4 production rate of 371 ± 100 L/(kgVS_{fed}·d) and a CH_4 concentration of *ca.* 60% when dissolved CO_2 levels inside the test reactor were increased up to 1.9-fold. Results also indicated an improvement in process resilience to temporary overloads and no impacts on stability parameters.

Keywords: anaerobic digestion; carbon dioxide utilisation; sewage sludge; pilot scale; process stability.

1. Introduction

Anaerobic digestion (AD) has recently been proposed as a promising system to biochemically convert exogenous carbon dioxide (CO_2) into methane (CH_4) (Bajón Fernández *et al.*, 2014; Salomoni *et al.*, 2011) and this option is finding growing interest thanks to the possibility of developing carbon negative renewable energy production (Cheah *et al.*, 2016; Budzianowski, 2012). CO_2 reduction to CH_4 in the AD process is traditionally associated with the activity of hydrogenotrophic methanogens (Demirel

ACCEPTED MANUSCRIPT

and Scherer, 2008). Homoacetogens can also play a role in reducing CO_2 and H_2 into acetic acid that is then transformed into CH_4 by acetoclastic methanogens (Liu *et al.*, 2016) or through syntrophic acetate oxidation followed by hydrogenotrophic methanogenesis (Schnürer and Nordberg, 2008). Whilst the biochemical mechanisms for exogenous CO_2 bioconversion in AD have not been fully elucidated, various authors have assessed the possibility to enhance CH_4 production from AD by CO_2 enrichment. Alimahmoodi and Mulligan (2008) studied, at lab scale, the possibility of converting CO_2 into CH_4 by using an up-flow anaerobic sludge blanket (UASB) reactor fed with a solution composed of dissolved CO_2 and volatile fatty acids (VFAs). The same authors observed a 69–86% CO_2 uptake, reporting that VFAs were used as source of H_2 for hydrogenotrophic methanogens to perform the CO_2 conversion to CH_4. Salomoni *et al.* (2011) studied at pilot scale the injection of CO_2 into the fermentation phase of a two-phase anaerobic digestion (TPAD) plant. Off gases from the fermentation phase were recirculated into the methanogenic phase to sustain CO_2 reduction to CH_4 and a 25% increase in CH_4 yield was observed. Similarly, Yan *et al.* (2016) studied the recirculation of off-gases from a TPAD reactor for food waste digestion. These authors utilised an acidogenic leach bed reactor, as first phase, and diverted off-gases (rich in CO_2 and H_2) and leachate from this reactor into a methanogenic UASB, used as second digestion phase. Results indicated an improvement of CH_4 production thanks to CO_2 and H_2 conversion to CH_4 that was assumed to be carried out by hydrogenotrophic methanogens.

These results highlight the biological feasibility of CO_2 bioconversion into CH_4 even though most of the studies utilised exogenous H_2 to support this bioprocess. The current lack of an inexpensive H_2 supply system and the low water solubility of H_2 are

ACCEPTED MANUSCRIPT

challenges that hinder the full exploitation of CO_2 bioconversion into CH_4 at AD sites by the use of exogenous H_2 (Bassani *et al.*, 2016). Similarly, the use of TPAD configuration could limit a large implementation of CO_2 bioconversion, considering that the majority of AD assets are single phase plants (De Baere and Mattheeuws, 2010). To overcome these limitations, an alternative approach could be based on the injection of CO_2 directly into digesters without any additional fermentation phase and without addition of exogenous H_2. Recent studies have assessed this procedure and indicated encouraging results. Bajón Fernández *et al.* (2014) studied the possibility to improve AD performance by direct CO_2 injection in single phase digestion, without the availability of exogenous H_2. Results from batch tests indicated an increase of CH_4 yields between 5 to 13% for food waste digestion and a speed up of CH_4 production for sewage sludge leading to an increase of *ca.* 100% on CH_4 production within the first 24 h of digestion, if compared to control experiments. A positive influence of exogenous CO_2 on AD performance during biochemical methane potential (BMP) tests was also reported by Koch *et al.* (2015; 2016), that observed an increase of CH_4 yields proportional to the CO_2 concentration of gases used to flush reactors head space. The benefit of direct injection of CO_2 on AD was also observed at pilot scale for food waste digestion (Bajón Fernández *et al.*, 2015). Results from this investigation indicated a 2.5-fold increase in H_2 concentration in the digester enriched with CO_2, that could support the conversion of exogenous CO_2 into CH_4, and resulted in a *ca.* 20% higher CH_4 production when comparing performance of test reactor before and after CO_2 injection. These results therefore support that biochemical conversion of exogenous CO_2 to CH_4 can be obtained in AD also without external supplementation of H_2. This option opens the possibility to exploit such biological process in various industrial sectors where AD

ACCEPTED MANUSCRIPT

is already an implemented technology. This could be further facilitated by the growing application of biogas upgrading to biomethane (Sun *et al.*, 2015) that is leading to the large availability of CO_2, directly on the digestion sites, that can be converted into CH_4, as promising approach to convert a waste stream into a commodity (Koch *et al.*, 2016). Enhancement of CH_4 production from sewage sludge AD supplemented with exogenous CO_2 has only been proved at batch scale (Bajón Fernández *et al.*, 2014) and further confirmations at larger scale are needed to proof the concept and clarify the long-term impacts of CO_2 injection on AD performance and stability. This research was therefore aimed at assessing, at pilot scale, the effects of exogenous CO_2 injection on single phase continuous AD of sewage sludge, without exogenous H_2 addition. The research focused on understanding the impacts of moderate and intense exogenous CO_2 injections on CH_4 production, biogas quality and AD process stability parameters.

2. Material and methods

2.1. Reactors configuration and operation

Two identical pilot scale AD reactors were used for the research study. The reactor used for CO_2 enrichment is hereafter referred to as Test reactor while the other is referred to as Control reactor. A scheme of the experimental rig is presented in Figure 1. Each unit was composed of a cylindrical reactor with a cone base having a total volume of 165 L. Working liquid volume was set to 90 L. Mixing of digestion material was performed by an external peristaltic pump (series 600, Watson Marlow, Cornwall, UK). Pump rate was set to have a full recirculation of the working liquid volume in 30 minutes. The AD process was carried out at mesophilic conditions. Temperature of digestion liquid was

ACCEPTED MANUSCRIPT

maintained at 38.5 ± 1 °C by using heating jackets (LMK Thermosafe, Haverhill, UK) placed over the cylindrical section of each reactor.

The reactors were operated semi-continuously with feeds carried out once a day. The feeding regime was repeated weekly as follows: 6 L of sewage sludge from the 1st to the 4th day of the week, 12 L of sewage sludge on the 5th day and no feed on the 6th and 7th day of the week. Micronutrients were added during any feed at a dosing rate of 0.05 mL of TEA 310 solution (Omex Environmental Ltd., King's Lynn, UK) per kg of volatile solids (VS) fed. The pH of feeding sewage sludge was not adjusted. The weekly average Hydraulic Retention Time (HRT) was 17.5 d and the average Organic Loading Rate (OLR) was 2.1 ± 0.4 kgVS/m^3·d. The two reactors were fed in parallel at the same time of the day and were maintained at the same feeding conditions for the entire experimental period.

The Test reactor was equipped with an external column retrofitted as a side process to perform the CO_2 enrichment of the digestion liquid. The column was connected to the Test reactor in the mixing loop only during each CO_2 enrichment (Figure 1). Test and Control reactors operated similarly during the rest of the time. No CO_2 injections were carried out on Test reactor until day 42.

Biogas production, biogas composition, pH and temperature of the digestion liquid were monitored five times per week. Samples of digestate from both reactors were collected up to 5 times a week to measure: Total Solid (TS), VS, Ammonium Nitrogen (NH_4^+), Partial Alkalinity (PA), Intermediate Alkalinity (IA), Total Alkalinity (TA), H_2CO_3 Alkalinity and total Volatile Fatty Acids (VFAs) concentration. The following single VFAs were also monitored: acetic acid, propionic acid, butyric acid and valeric acid.

ACCEPTED MANUSCRIPT

120 **2.2. Feeding material and inoculum of reactors**

121 Sewage sludge was used as feedstock for the reactors. The sewage sludge used in this
122 study was a mixture of primary sludge and waste activated sludge produced in a
123 municipal wastewater treatment works (WwTW) located in the Midlands area of UK.
124 Sludge was collected from the inlet flow of a full-scale AD plant located in this
125 WwTW. After collection, samples were stored at 4 °C until use. Four batch samples of
126 sludge were collected at different times during the experiment and are named Sample 1,
127 Sample 2, Sample 3 and Sample 4. During the entire experiment, both reactors were fed
128 with the same sludge sample. Phases of the experiment during which the four samples
129 of sludge were used are reported in Figures 2, 3, 5 and 6.
130 The composition of each sample of sludge was monitored for the following parameters:
131 TS, VS, NH_4^+, TA, H_2CO_3 alkalinity, total and single (acetic acid, propionic acid,
132 butyric acid and valeric acid) VFAs concentration. Average characteristics of each
133 sample are reported in Table 1.
134 Reactors were inoculated with digestate collected from a full-scale mesophilic
135 anaerobic digester located in the same WwTW. TS and VS concentrations of the
136 inoculum were 30 ± 2 gTS/L and 18 ± 1 gVS/L, respectively.
137

138 **2.3. Carbon dioxide injection procedure**

139 CO_2 enrichment of digestion liquid was performed by using a 1 m tall and 10 cm
140 diameter column located in the recirculation loop of the Test AD reactor (Figure 1). The
141 column was operated with a liquid working volume of 7 L. CO_2 was injected at the
142 bottom of the column through a perforated plate. A metallic mesh with 0.5 mm hole size
143 was placed on top of the perforated plate to generate small gas bubbles enhancing CO_2

ACCEPTED MANUSCRIPT

144 dissolution into the digestion liquid. The contact between digestion liquid and CO_2 was
145 performed in co-current mode.
146 In order to assess the impact of dissolved CO_2 levels in AD operation, two different
147 column configurations were used. The first was a bubble column configuration with
148 internal space of the column empty. The second was a packed column configuration in
149 which the internal space was filled with small perforated plastic media of cylindrical
150 shape and various dimensions (length = 5 cm, diameters = 1, 2 and 4 cm) having
151 rectangular openings of *ca.* 2 x 10 mm evenly distributed on the surface.
152 The moderate CO_2 enrichment was carried out between day 42 and day 76, with three
153 CO_2 injections per week using the bubble column configuration. The intense CO_2
154 enrichment was performed between day 91 and day 127 with five CO_2 injections per
155 week using the packed column configuration. Between these two phases, Test rector
156 was operated without CO_2 injection for 14 days.
157 During both phases, the CO_2 injection was carried out for 1 hour at a time maintaining a
158 fixed CO_2 flow rate into the column of 1.5 L/min by means of a mass flow controller
159 (MFC) (Premier Control Technologies, Norfolk, UK). CO_2 was supplied from gas
160 cylinders (BOC, Manchester, UK). The mixing pump speed was reduced during
161 injection in order to increase the gas to liquid contact time in the column and to
162 circulate the entire digestion liquid through the column during the 1-hour operation. The
163 same speed reduction was applied to the mixing pump of the Control reactor for the
164 length of the CO_2 injection procedure. CO_2 enrichment was performed at the same time
165 of the day and always before feeding both the reactors. The experimental set up used
166 was similar to the one reported by Bajón Fernández *et al.* (2015).

ACCEPTED MANUSCRIPT

Dissolved CO_2 concentration and pH were measured in the digestion liquid of the Test reactor at the beginning and at the end of any CO_2 enrichment, while dissolved CO_2 concentration and pH of the liquid entering and exiting the CO_2 injection column were measured every 10 minutes. Concentrations of CO_2 and CH_4 in the column gas exhaust (Figure 1) were measured every 5 minutes. At the end of any CO_2 enrichment, biogas composition in the Test reactor head space was also measured.

2.4. Analytical methods and statistical analysis

Biogas production was measured by drum-type gas meters (Ritter TG 05/5, Germany). Biogas composition was measured by means of a portable gas analyser (LMSXi multifunction gas analyser, Gas Data, Coventry, England) and data on biogas mixing ratio are reported as concentrations expressed in %. Dissolved CO_2 concentrations were measured by means of CO_2 sensors (InPro®5000(i), Mettler-Toledo AG, Switzerland) connected to a multiparameter transmitter (M400, Mettler-Toledo AG, Switzerland). Concentrations of CO_2 and CH_4 in the column gas exhaust (Figure 1) were measured by means of gas sensors (BCP sensors, Bluesens, Herten, Germany) and recorded in a computer using BacVis software (Bluesens, Herten, Germany).

TS and VS were measured on raw samples according to Standard Methods (APHA, 2005). NH_4^+, IA, PA, TA, H_2CO_3 alkalinity and total and single VFAs, were measured on the supernatant of samples centrifuged for 20 minutes at 8000 g and 20 °C. NH_4^+ was quantified by using Spectroquant test kits (Merck, Germany). Alkalinities and total VFAs were measured by titration with 0.06 N HCl acid on supernatants diluted 1:10 in deionised water. IA and PA were measured by titration to pH values of 5.75 and 4.30, respectively, and IA/PA ratio was calculated as ratio between titration volumes (Ripley

ACCEPTED MANUSCRIPT

191 *at al.*, 1986). TA, H_2CO_3 alkalinity and total VFAs were measured by titration at 8 pH
192 points as reported by Lahav *et al.* (2002). The ratio between total VFAs and H_2CO_3
193 alkalinity measured by this titration procedure is referred as VFA/Alk ratio in the
194 present study.
195 To measure single VFAs, supernatants were filtered through 0.45 µm pore size syringe-
196 drive filters (MilliporeTM, Billerica, United States). High performance liquid
197 chromatography (HPLC) (Shimadzu VP Series unit, Milton Keynes, UK) was utilised
198 to quantify concentration of acetic acid, propionic acid, butyric acid and valeric acid.
199 The methodology is reported in Soares *et al.* (2010) with the only exception that a
200 HPLC run time of 60 minutes was used in this research.
201 Results from both Control and Test reactors were statistically evaluated by means of
202 sign test. Sign test is a non-parametric test with dependent samples ordered in pairs. A
203 confidence level of 95% was selected for all statistical comparisons.
204
205 **3. Results and Discussion**
206 **3.1. Sewage sludge digestion performance and effects of CO_2 injection**
207 A comparison of Control and Test reactors performance during the different phases of
208 the experimental work is presented in Table 2. Control and Test reactors are compared
209 for results before the CO_2 injection started and during the two phases of CO_2 injection
210 performed at different frequencies and column configurations. Trends of CH_4 and H_2
211 concentrations for the entire experimental period are reported in Figure 2. Trends of
212 NH_4^+ concentration in digestate and H_2CO_3 alkalinity are presented in Figure 3a while
213 pH trends are presented in Figure 3b. Figure 4 presents the average change in pH on
214 digestate exiting the injection column during CO_2 enrichment and the average increase

ACCEPTED MANUSCRIPT

in dissolved CO_2 concentration compared to the starting point (C/C_0). The final C/C_0 achieved in the Test AD after completing the CO_2 injection is also reported.

The Control reactor showed unstable performance during the first two weeks (data not shown), therefore it was reseeded and feeding started again, at the same feed rate of Test reactor, on day 19. From day 19 onwards, both reactors showed stable operational conditions with similar process performance ($p>0.05$). During the period without CO_2 enrichment (first 42 days) average CH_4 concentration was 65 ± 3% for both reactors (Table 2 and Figure 2) and specific CH_4 production was 373 ± 169 and 384 ± 175 $L/(kgVS_{fed} \cdot d)$ for Control and Test reactors, respectively (Table 2), H_2 concentrations followed similar patterns with a slight increase in concentration after day 30 for both reactors (Figure 2).

The decreasing trend of H_2CO_3 alkalinity (Figure 3a) was probably due to a change in organic nitrogen content of feed sludge as also indicated by the decreasing trend of NH_4^+ concentration in the reactors. Degradation of organic nitrogen to NH_4^+ is in fact the main way in which alkalinity is generated during biodegradation of organic matter (Rittmann and McCarty, 2001). IA/PA and VFA/Alk ratio remained below 0.4 and 0.2, respectively (Figure 5). Acetic and propionic acids showed similar trends for both Test and Control reactors with no peaks in concentration (Figure 6a) during the initial phase of the research without CO_2 enrichment, indicating a stable operational condition.

Overall, the differences between monitoring parameters (Table 2) did not result statistically different ($p>0.05$).

The first phase of CO_2 injection started on Test reactor on day 42, with 3 injections per week by means of a bubble column.

ACCEPTED MANUSCRIPT

238 The dissolution of a weak acid during CO_2 enrichment produced a temporary reduction
239 in pH and this effect can be observed on the decreasing trend of pH in the effluent from
240 the injection column (Figure 4a and 4b). On average, the use of a bubble column
241 (Figure 4a) produced a pH reduction of about 0.10 points while injections with a packed
242 column (Figure 4b) reduced the pH by 0.15 points. The use of a packed column in fact
243 allowed a higher CO_2 dissolution, as confirmed by the higher C/C_0 ratio reached during
244 the second phase of CO_2 injection (Figure 4b).
245 Both reactors showed a decreasing trend of pH (Figure 3b) that can be associated to the
246 reduction in organic nitrogen content on feed sludge as confirmed by the lowering
247 pattern of NH_4^+ concentrations (Figure 3a), as already discussed. The Test reactor did
248 not show any additional decreasing trend of pH during CO_2 enrichment, indicating that
249 the system was able to recover after the temporary pH reduction in digestion liquid
250 exiting the column. CO_2 injection did not impact therefore H_2CO_3 alkalinity of the Test
251 reactor (Figure 3a). These results confirm observations reported by Bajón Fernández *et*
252 *al.* (2014) where CO_2 enrichment of batch tests treating sewage sludge and food waste
253 indicated that the initial acidification associated with CO_2 injection was overcome
254 within one day. Bajón Fernández *et al.* (2015) during pilot scale digestion of food waste
255 did not observe a reduction on digestion pH with a CO_2 enrichment frequency of 3
256 injections per week, similarly to the moderate frequency on the present study. Al-
257 mashhadani *et al.* (2016) also indicated a short-term effect of pH reduction during CO_2
258 injection, followed by a recovery phase when injection was not performed, in a gaslift
259 digester sparged with pure CO_2 for 5 minutes a day. An overall increasing pH trend was
260 also observed for this reactor, but a comparison with a control unit was not reported.

ACCEPTED MANUSCRIPT

These results therefore suggest that the CO_2 enrichment procedure has no long term impacts on pH under continuous operating conditions.

During the first phase of CO_2 injection, a variable H_2 concentration for Test reactor was observed, with peaks up to 220 ppm (Figure 2 and Table 2). On the contrary H_2 concentration for Control reactor remained stable at values close to 110 ppm from day 42 onwards. In the first phase of CO_2 injection, CH_4 concentration in Test reactor resulted rather variable (Figure 2). Average concentration for Test reactor was 59 ± 3% while for Control reactor was 62 ± 2% ($p<0.05$) (Table 2). During the second phase of CO_2 injection, started on day 92 with 5 injections per week and a packed column configuration, H_2 concentration of Test reactor showed a higher average concentration ($p<0.05$) than the Control, 138 ± 26 ppm and 107 ± 10 ppm, respectively, and average CH_4 concentration was slightly lower ($p<0.05$), with an average of 61 ± 2% and 63 ± 2% in Test and Control reactors, respectively (Table 2).

An increasing concentration of H_2 in biogas together with growing concentrations of organic acids in digestate is typically reported as an indicator of overloading or inhibitory conditions for anaerobic bioreactors (Voolapalli and Stuckey, 2001; Ketheesan and Stuckey, 2015). Accumulation of intermediates indicates in fact an unbalanced condition between the activity of acetogens and methanogens due to a fast change of process conditions. The peaks in H_2 concentration observed after the start of CO_2 injection, could be associated to a release of protons when carbonic acid dissociates into carbonate and bicarbonate (Bajón Fernández *et al.*, 2015) but could also suggest that this procedure introduced a disturbance in the biological process affecting the activity of H_2 consuming microorganisms or be related to a boost of H_2 producing metabolisms. Increase of H_2 concentration due to a reduction of hydrogenotrophic

ACCEPTED MANUSCRIPT

activity is usually simultaneous to increases of propionate or butyrate acids due to syntrophic degradation of these intermediates (Voolapalli and Stuckey, 2001). As no reduction of biogas or CH_4 production (Table 2) or indications of process instability were recorded, it is likely that the increase of H_2 production and of these acids was associated to an increased acidogenic activity stimulated by the CO_2 injection rather than an inhibition of hydrogenotrophic activity. No clear trends of VFA concentration were anyway observed, suggesting that further work is needed to elucidate the mechanisms of utilization of the injected CO_2. The CO_2 injection, both at moderate and intense frequency, did not lead to increasing levels of H_2, but to a new H_2 baseline which, for Test reactor, stabilised at *ca.* 138 ppm (Table 2). The fact that the H_2 concentration reached a new baseline rather than maintaining an increasing trend, suggests that hydrogenotrophic activity was stimulated because of a higher substrate availability. Bajón Fernández *et al.* (2015) also measured an increasing trend of H_2 concentration with a new baseline being reached at 320 ± 153 ppm in biogas during CO_2 enrichment of a pilot scale food waste AD. In that study, the higher H_2 production was attributed to either a chemical process of proton formation due to CO_2 dissolution into carbonate/bicarbonate, or to a biologically enhanced acetogenesis. The increased H_2 consumption (new H_2 baseline rather than a rising trend) was in this case attributed to a potential increase in homoacetogenesis via the Wood-Ljungdahl pathway (Bajón Fernández *et al.*, 2015). Al-mashhadani *et al.* (2016) suggested that the addition of CO_2 in an anaerobic gaslift bioreactors of kitchen waste, deploying microbubbles generated by fluidic oscillation, could boost H_2 production (and consequently CH_4 production) due to an improved hydrolysis of organics given by the collapse of microbubbles generating radicals able to facilitate the disruption of slowly biodegradable organics.

ACCEPTED MANUSCRIPT

This hypothesis could explain both the higher H_2 concentration observed during the experimental period and the increased CH_4 production (Table 2). The injection of CO_2 could therefore increase H_2 levels as a result of improved hydrolysis but this assumption needs further confirmation as the equipment utilised in this research study was not designed to generate microbubbles.

CH_4 production resulted differently affected during the two injection phases (moderate and intense) (Table 2). In the first phase, characterised by 3 injections per week with a bubble column, average specific CH_4 productions resulted similar. During the intense phase of CO_2 injection, 5 injections per week with a packed column, average specific CH_4 production in the Test reactor (371 ± 100 L/($kgVS_{fed}$·d)) was *ca.* 12% higher than for the Control Reactor (332 ± 94 L/($kgVS_{fed}$·d)) and in this case productions over time were statistically different (paired sign test, $p<0.05$). The increase in CH_4 production could be explained by an increased hydrogenotrophic methanogenesis, by an increased acetoclastic methanogenesis or by an increased methylotrophic methanogenesis. An increased hydrogenotrophic methanogenesis could be a result of a stimulation of H_2 production pathways as a response to the increased inorganic carbon availability, as previously described, while a boost in acetate availability because of utilisation of CO_2 in the Wood-Ljungdahl mechanism can explain an increase in activity of acetoclastic methanogens leading to higher CH_4 productions (Bajón Fernández *et al.*, 2015). The reduction of exogenous CO_2 and H_2 to methanol is also another possible route for higher CH_4 production that is linked to conversion of methanol to CH_4 by methylotrophic methanogens (Guo *et al.*, 2015).

A higher CH_4 production was also observed by Salomoni *et al.* (2011) during CO_2 injection on TPAD of sewage sludge at pilot scale. These authors achieved a 25%

ACCEPTED MANUSCRIPT

increase in CH_4 production, if compared to a full-scale single phase digestion plant, by injecting CO_2 into the acidogenic stage of the TPAD process. In the present study, the improvement of CH_4 production associated with CO_2 enrichment was *ca.* 12%. Even though the two systems have similar HRTs (~17 d), differences as OLR (1.05 ± 0.04 vs. 2.1 ± 0.4 kgVS/m^3·d in the present study), plant configuration (double phase vs. single phase in the present study), injection procedure (continuous vs. intermittently in the present study), and the specific conditions of the digestion liquid during injection (acidic vs. neutral-alkaline in the present study) limit the comparability of results. Enhancement of CH_4 production was also reported by Al-mashhadani *et al.* (2016) during pure or diluted biogas recirculation, or CO_2 injection in anaerobic gaslift bioreactors of kitchen waste, using microbubbles generated by fluidic oscillation. These authors described that the injection of recirculated biogas (with CO_2 concentration of 40 or 80%) increased CH_4 production between 10 and 14% while the injection by microbubbles of pure CO_2 increased CH_4 production by more than 100%. It is suggested that this procedure stimulates CH_4 production due to two processes. The first is a faster removal of CH_4 from the liquid phase that reduces its partial pressure and thermodynamically enhances reactions having CH_4 as final product. The second process links the higher CH_4 production to an increased hydrolysis. In the present study, no net difference was recorded on VS concentrations between the two reactors ($p>0.05$) (Table 2) therefore it is not possible to confirm an improved solids degradation even though CH_4 production was higher with injection of CO_2, if compared to Control reactor. Further studies are therefore necessary to gain a better understanding of this aspect.

3.2 Anaerobic digestion process stability under CO_2 injection

ACCEPTED MANUSCRIPT

Variations over time of process stability parameters (IA/PA and VFA/Alk ratios) are reported in Figure 5. Concentrations of acetic and propionic acids are reported in Figure 6a, concentrations of butyric and valeric acids are reported in Figure 6b.

During the first 42 days in which both reactors were maintained at the same loading conditions and CO_2 injection was not performed on Test reactor, IA/PA and VFA/Alk parameters remained within ranges indicating good stability of the biological process, (IA/PA < 0.4 and VFA/Alk < 0.2, Li *et al.*, 2014; Vannecke *et al.*, 2014) and VFAs concentration showed comparable trends between the two reactors (Figures 5 and 6). From day 42, both reactors showed some peaks of both IA/PA and VFA/Alk ratios. Control reactor showed peaks of these parameters on day 50, 65 and 108. On day 50, IA/PA and VFA/Alk ratios for Control reactor reached values of 0.55 and 0.6, respectively, while during the other two events IA/PA ratio resulted close to or higher than 0.5 and VFA/Alk ratio higher than 0.3. Test reactor also showed peaks of these parameters on the same days, but the increase resulted less intense (Figure 5). During the moderate phase of CO_2 injection characterised by 3 injections per week, IA/PA ratio of the Test reactor reached peaks of about 0.45 on days 50 and 65, while VFA/Alk ratio increased to values of about 0.25 on the same days. During the intense phase of the injection procedure, characterised by 5 injections per week, results from the Test reactor indicated that IA/PA never exceeded 0.4 and VFA/Alk remained stable around 0.1 (Figure 4).

Observing the trends of concentration of VFAs (Figure 6a and 6b), an increase in acetic, propionic and butyric acids was recorded during the days in which peaks in stability parameters (IA/PA, VFA/Alk) were measured. Similarly, a reduction of H_2CO_3 alkalinity was also observed during these events (Figure 3a).

ACCEPTED MANUSCRIPT

As these variations in process parameters were observed for both reactors, it is presumable that they were a response to a temporary unbalanced process condition caused by a change of feeding load or composition. Even though reactors were fed with the same volume of sewage sludge (see paragraph 2.1), variations in solids concentrations and sludge composition over time could have imposed changes on loading rates on reactors. Both reactors recovered quickly from these temporary unbalanced conditions without requiring any reduction in feeding regime. However, it is of note that the Test reactor showed lower peaks of stability parameters than the Control reactor during all these events, while it was subjected to CO_2 enrichment. In fact, IA/PA and VFA/Alk ratios for the Test reactor never exceed 0.45 and 0.25, respectively, in all these occasions, while the Control reactor reached values of IA/PA and VFA/Alk ratios up to 0.55 and 0.6, respectively. Similarly, acetic acid concentrations in the Test reactor resulted always lower than those for Control reactor (Figure 6a).

This increased resilience of the Test reactor is particularly evident during the third event around day 105. IA/PA and VFA/Alk ratios remained at high values for about 10 days for the Control reactor, while only small variations were recorded for the same parameters for the Test reactor (Figure 5). Acetic acid concentrations also remained above 500 mg/L for about ten days in the Control reactor, while a moderate peak, below 500 mg/L, and a fast recovery, less than 5 days, was observed for the Test reactor (Figure 6a).

These observations suggest that the injection of CO_2 on Test reactor induced a higher resilience to temporary overloads caused by sudden variations of feed composition at constant volumetric loads. Improved resilience as an effect of CO_2 injection was also observed by Bajón Fernández *et al.* (2015) during anaerobic digestion of food waste at

ACCEPTED MANUSCRIPT

pilot scale. The AD reactor enriched with CO_2 faced a sudden temperature drop of 12.5 °C that caused a decrease of both biogas production and pH. No VFA accumulation was observed and the reactor recovered from the stress condition much faster than the Control reactor, subject to a similar temperature drop, which required a partial re-seed to recover. No other studies have investigated the effect of CO_2 injection on AD process resilience, but the similar results obtained in this research study and by Bajón Fernández *et al.* (2015) observed from different stress conditions, suggest that the CO_2 enrichment procedure not only can be applied to boost CH_4 production but also can enhance process stability and resilience.

The higher resilience observed for the Test reactor could be associated with a higher heterogeneity or functional redundancy of microbial populations within the process stimulated by CO_2 enrichment. A more diversified microbial community expressing a high degree of redundancy for trophic pathways, is suggested to maintain a high rate of degradation activity and process stability even under variability of feed composition or organic load (Briones and Raskin, 2003). This could explain why stability parameters showed lower peaks and faster recovery for the Test reactor in this study. Strategies to control or recover digesters from hydraulic or loading shock currently focus on stimulating either methanogenic activity or propionate and butyrate consumption by microbial bioaugmentation in an attempt to maximise intermediate consumptions and speed up process recovery (Ketheesan and Stuckey, 2015). Lerm *et al.* (2012) indicated that the coexistence of hydrogenotrophic and acetoclastic methanogens is necessary to respond to process perturbations and leads to stable process performance during shock load conditions. Shifts from acetoclastic to hydrogenotrophic methanogens were in fact

ACCEPTED MANUSCRIPT

reported during organic overloads as a response to high H_2 availability (Lerm *et al.*, 2012).

From an overall point of view, CO_2 injection did not produce negative impacts on biological stability of the Test reactor. Excluding the three events during which an overload of both reactors was observed, IA/PA and VFA/Alk remained within values normally reported for stable performance (Ketheesan and Stuckey, 2015; Ripley *at al.*, 1986). No accumulation of VFAs was observed during both moderate and intense CO_2 enrichment phases (Figure 6). On the contrary, average acetic acid concentration in the Test reactor (200 ± 120 mg/L) resulted lower than in the Control reactor (320 ± 180 mg/L) and butyric acid concentration in the Test reactor remained below concentrations measured in the Control reactor, in particular during the second (intense) phase of CO_2 injection (Figure 6b). These observations further support the hypothesis that a higher CH_4 production could be a result of an increased acidogenic activity. These results also suggest that the implementation of CO_2 enrichment in full scale AD operations can improve process resilience and potentially accommodate extra-loading capacity. Moreover, CO_2 enrichment could potentially represent a controlling strategy for digestion plants in which feed composition variability can easily create overloading conditions and inhibit the biological process. Further studies are required to understand whether CO_2 enrichment can enable an increased process capacity by supporting stable operation at higher OLR and lower HRT in single-phase continuous digestion processes.

4. Conclusions

ACCEPTED MANUSCRIPT

This study confirmed at pilot scale the possibility to enhance AD of sewage sludge by CO_2 enrichment without exogenous H_2 addition. The injection of exogenous CO_2 into AD represents a promising option to improve CH_4 production in a single-phase digester. Specific CH_4 production was increased by *ca.* 12% and no impacts were observed on the AD stability parameters that remained within typical ranges. CO_2 enrichment also allowed an increased process resilience to temporary overloads. CO_2 enrichment of sludge ADs has potential to enable a carbon-negative sewage sludge management with limited changes in process operation and control.

Acknowledgements

The authors gratefully acknowledge the support and funding provided by Severn Trent Water.

ACCEPTED MANUSCRIPT

ACCEPTED MANUSCRIPT

464 **REFERENCES**

465 1. Al-mashhadani, M.K.H., Wilkinson, S.J., Zimmerman, W.B. 2016. Carbon dioxide
466 rich microbubble acceleration of biogas production in anaerobic digestion. Chem Eng
467 Sci 156, 24–35.
468 2. Alimahmoodi, M., Mulligan, C.N., 2008. Anaerobic bioconversion of carbon dioxide
469 to biogas in an upflow anaerobic sludge blanket reactor. J Air Waste Manage. 58, 95–
470 103.
471 3. APHA-AWWA-WEF., 2005. Standard Methods for the Examination of Water and
472 Wastewater, 21st ed., American Public Health Association, American Water Works
473 Association, Water Environment Federation. Washington, D.C.
474 4. Bajón Fernández, Y., Soares, A., Villa, R., Vale, P., Cartmell, E. 2014. Carbon
475 capture and biogas enhancement by carbon dioxide enrichment of anaerobic digesters
476 treating sewage sludge or food waste. Bioresource Technol. 159, 1–7.
477 5. Bajón Fernández, Y., Green, K., Schuler, K., Soares, A., Vale, P., Alibardi, L.,
478 Cartmell, E. 2015. Biological carbon dioxide utilisation in food waste anaerobic
479 digesters. Water Res 87, 467-475.
480 6. Bassani, I, Kougias, P.G., Angelidaki, I., 2016. In-situ biogas upgrading in
481 thermophilic granular UASB reactor: key factors affecting the hydrogen mass transfer
482 rate. Bioresource Technol 221, 485–491
483 7. Briones, A., Raskin, L. 2003. Diversity and dynamics of microbial communities in
484 engineered environments and their implications for process stability. Curr Opin
485 Biotech. 14, 270-276.
486 8. Budzianowski, W.M. 2012 Negative carbon intensity of renewable energy
487 technologies involving biomass or carbon dioxide as inputs. Renew Sust Energ Rev.
488 16, 6507-6521.

ACCEPTED MANUSCRIPT

9. Cheah, W.Y., Ling, T.C., Juan, J.C., Lee, D-J., Chang, J-S., Show, P.L. 2016. Biorefineries of carbon dioxide: From carbon capture and storage (CCS) to bioenergies production. Bioresource Technol. 215, 346–356

10. De Baere, L., Mattheeuws, B., 2010. Anaerobic digestion of MSW in Europe, 2010 update and trends. Biocycle, February 2010, pp. 24-26

11. Demirel, B., Scherer, P. 2008. The roles of acetotrophic and hydrogenotrophic methanogens during anaerobic conversion of biomass to methane: a review. Rev Environ Sci Bio. 7, 173–190.

12. Guo, J., Peng, Y., Ni, B.J., Han, X., Fan, L., Yuan, Z., 2015. Dissecting microbial community structure and methane-producing pathways of a full-scale anaerobic reactor digesting activated sludge from wastewater treatment by metagenomics sequencing. Microb Cell Fact, 14, 33-44.

13. Lahav, O., Morgan, B. E., Loewenthal, R. E., 2002. Rapid, simple, and accurate method for measurement of VFA and carbonate alkalinity in anaerobic reactor. Environ Sci Technol. 36, 2736-2741.

14. Lerm, S., Kleyböcker, A., Miethling-Graff, R., Alawi, M., Kasina, M., Liebrich, M., Würdemann, H., 2012. Archaeal community composition affects the function of anaerobic co-digesters in response to organic overload. Waste Manage 32, 389–399

15. Li, L., He, Q., Wei, Y., He, Q., Peng, X. 2014. Early warning indicators for monitoring the process failure of anaerobic digestion system of food waste. Bioresource Technol 171, 491-494

16. Liu, R., Hao, X., Wei, J. 2016. Function of homoacetogenesis on the heterotrophic methane production with exogenous H_2/CO_2 involved. Chem Eng J. 284, 1196–1203

ACCEPTED MANUSCRIPT

17. Ketheesan, B., Stuckey, D.C., 2015. Effects of Hydraulic/Organic Shock/Transient Loads in Anaerobic Wastewater Treatment: A Review. Crit Rev Env Sci Tec. 45, 2693-2727

18. Koch, K., Bajón Fernández Y., Drewes J.E. 2015. Influence of headspace flushing on methane production in Biochemical Methane Potential (BMP) tests. Bioresource Technol. 186, 173–178

19. Koch, K., Huber, B., Bajón Fernández, Y., Drewes, J.E. 2016. Methane from CO2: Influence of different CO2 concentrations in the flush gas on the methane production in BMP tests. Waste Manage. 49, 36–39

20. Ripley, L. E., Boyle, W. C., Converse, J. C., 1986. Improved alkalimietric monitoring for anaerobic digestion of high-strenght wastes. J Water Pollut Con F - WPCF. 58, 406-411.

21. Rittmann, B. E., McCarty P. L., 2001. Environmental Biotechnology: Principles and Applications. McGraw-Hill ISBN 0-07-118184-9

22. Salomoni, C., Caputo, A., Bonoli, M., Francioso, O., Rodriguez-Estrada, M.T., Palenzona, D., 2011. Enhanced methane production in a two-phase anaerobic digestion plant, after CO_2 capture and addition to organic wastes. Bioresource Technol. 102, 6443-6448.

23. Schnürer, A., Nordberg, A., 2008. Ammonia, a selective agent for methane production by syntrophic acetate oxidation at mesophilic temperature. Water Sci Technol. 57, 735-740.

24. Soares, A., Kampas, P., Maillard, S., Wood, E., Brigg, J., Tillotson, M., Parsons, S.A., Cartmell, E., 2010. Comparison between disintegrated and fermented sewage sludge for production of a carbon source suitable for biological nutrient removal. J Hazard Mater. 175, 733–739.

ACCEPTED MANUSCRIPT

537 25. Sun, Q., Li, H., Yan, J., Liu, L., Yu, Z., Yu, X. 2015. Selection of appropriate biogas
538 upgrading technology-a review of biogas cleaning, upgrading and utilisation. Renew
539 Sust Energ Rev. 51, 521–532
540 26. Vannecke, T.P.W., Lampens, D.R.A., Ekama, G.A., Volcke, E.I.P. 2014. Evaluation
541 of the 5 and 8 pH point titration methods for monitoring anaerobic digesters treating
542 solid waste. Environ Technol. 36, 861-869
543 27. Voolapalli, R., Stuckey, D.C. 2001. Hydrogen production in anaerobic reactors during
544 shock loads: influence of formate production and H_2 kinetics. Water Res. 35, 1831–
545 1841.
546 28. Yan, B.H., Selvam, A. Wong, J.W.C. 2016. Innovative method for increased methane
547 recovery from two-phase anaerobic digestion of food waste through reutilization of
548 acidogenic off-gas in methanogenic reactor. Bioresource Technol. 217, 3–9.

ACCEPTED MANUSCRIPT

ACCEPTED MANUSCRIPT

549 **Table caption**s

550

551 Table 1. Average physical and chemical composition of the samples of sewage sludge
552 used as feedstock. Temporal reference on when samples were used during the
553 experiments are reported in Figure 2, 3, 5 and 6.

554

555 Table 2. Average data (± Standard Deviation) obtained from the Control and Test
556 reactors during the different phases of the experimental period. Star (*) indicates
557 statistically different data ($p < 0.05$) between the same experimental condition.

558

559

560

561

562

563

564

565

566

567

568

569

ACCEPTED MANUSCRIPT

ACCEPTED MANUSCRIPT

570 **Table 1**. Average physical and chemical composition of the samples of sewage sludge
571 used as feedstock. Temporal reference on when samples were used during the
572 experiments are reported in Figure 2, 3, 5 and 6.

Parameter	**Sample 1**	**Sample 2**	**Sample 3**	**Sample 4**
Total Solids (%)	5.4	4.1	6	4.5
Volatile Solids (% of TS)	77	79	80	81
pH	6.06	5.61	5.51	5.98
NH_4^+ (mgN/L)	435	370	210	90
Total alkalinity (mg$CaCO_3$/L)	3500	5500	2900	4200
H_2CO_3 alkalinity (mg$CaCO_3$/L)	820	1180	600	1050
Total VFAs (mgCH_3COOH/L)	3100	4800	2600	3350
Acetic acid (mg/L)	500	1250	1500	1800
Propionic acid (mg/L)	800	2200	2800	1200
Butyric acid (mg/L)	620	1400	1600	1150
Valeric acid (mg/L)	900	1420	1900	3600

573

574

ACCEPTED MANUSCRIPT

ACCEPTED MANUSCRIPT

575 **Table 2.** Average data (± Standard Deviation) obtained from the Control and Test reactors during the different phases of the experimental

576 period. Star (*) indicates statistically different data ($p < 0.05$) between the same experimental condition.

	No CO_2 injection		3 CO_2 injections/week		5 CO_2 injections/week	
Parameter	**Control**	**Test**	**Control**	**Test**	**Control**	**Test**
pH	7.68 ± 0.08	7.69 ± 0.08	7.46 ± 0.08	7.38 ± 0.10	7.35 ± 0.06	7.28 ± 0.05
TS (g/L)	26.6 ± 2.3	27.8 ± 0.8	24.8 ± 0.7	24.6 ± 1.9	21.1 ± 2.2	22.0 ± 2.9
VS (g/L)	16.4 ± 1.5	17.1 ± 0.4	15.5 ± 0.5	15.6 ± 1.0	13.7 ± 1.1	13.8 ± 1.3
NH_4^+ (mgN/L)	1608 ± 124	1575 ± 141	1219 ± 220	1239 ± 131	944 ± 33	989 ± 61
Biogas production (L/d)	132 ± 35	141 ± 33	*119 ± 41	*140 ± 33	*126 ± 25	*147 ± 31
CH_4 production (L/d)	86 ± 24	91 ± 21	74 ± 26	83 ± 20	*80 ± 17	*90 ± 21
Specific CH_4 production (L/(kgVSfed·d))	373 ± 169	384 ± 175	290 ± 107	333 ± 112	*332 ± 94	*371 ± 107
CH_4 concentration (%)	65 ± 3	65 ± 3	*62 ± 2	*59 ± 3	*63 ± 2	*61 ± 2
H_2 concentration (ppm)	80 ± 23	72 ± 23	*113 ± 11	*126 ± 36	*107 ± 10	*138 ± 26

577

578

ACCEPTED MANUSCRIPT

ACCEPTED MANUSCRIPT

Figure captions

Figure 1. Scheme of the experimental rig. (a) Control reactor and (b) Test reactor configuration during CO_2 injection. (1) Anaerobic reactor, (2) heating jacket, (3) peristaltic pump, (4) biogas sample point, (5) biogas meter, (6) bubble column, (7) mass flow controller, (8) gas pressure regulator, (9) CO_2 cylinder, (10) CH_4-CO_2 analyser, (11) digestate sampling point.

Figure 2. Methane (CH_4) production, CH_4 and hydrogen (H_2) concentration in Test and Control reactors during the experimental period. Black vertical lines divide the phases of the experimental period between: no CO_2 injections phase (No CO_2 inj.), phase of moderate CO_2 enrichment at 3 injections per week with a bubble column (3 CO_2 inj./week) and phase of intense CO_2 enrichment at 5 injections per week with a packed column (5 CO_2 inj./week). Top grey line identifies when different samples of sludge were used.

Figure 3. Ammonium nitrogen (NH_4^+), H_2CO_3 Alkalinity concentrations (a) and pH (b) for Test and Control reactors during the different phases of the experimental period. Vertical lines divide the phases of the experimental period. Top grey line identifies when different samples of sludge were used.

Figure 4. Evolution of the parameter C/C_0 representing the ratio between the initial CO_2 concentration in digestate (C_0) and the concentration on the effluent of the CO_2 injection column (C). Evolution of pH in the effluent of the CO_2 injection column.

ACCEPTED MANUSCRIPT

The C/C_0 achieved in the Test reactor at the end of the injection is marked as "X". Graph a) is for the use of a bubble column, graph b) is for the use of a packed column.

Figure 5. Intermediate to Partial Alkalinity (IA/PA) ratio and volatile fatty acids to H_2CO_3 Alkalinity (VFA/Alk) ratio for Test and Control reactors during the different phases of the experimental period. Vertical lines divide the phases of the experimental period. Top grey line identifies when different samples of sludge were used.

Figure 6. Acetic and propionic acid concentrations (a) and butyric and valeric acid concentrations (b) for Test and Control reactors during the different phases of the experimental period. Vertical lines divide the phases of the experimental period. Top grey line identifies when different samples of sludge were used.

ACCEPTED MANUSCRIPT

ACCEPTED MANUSCRIPT

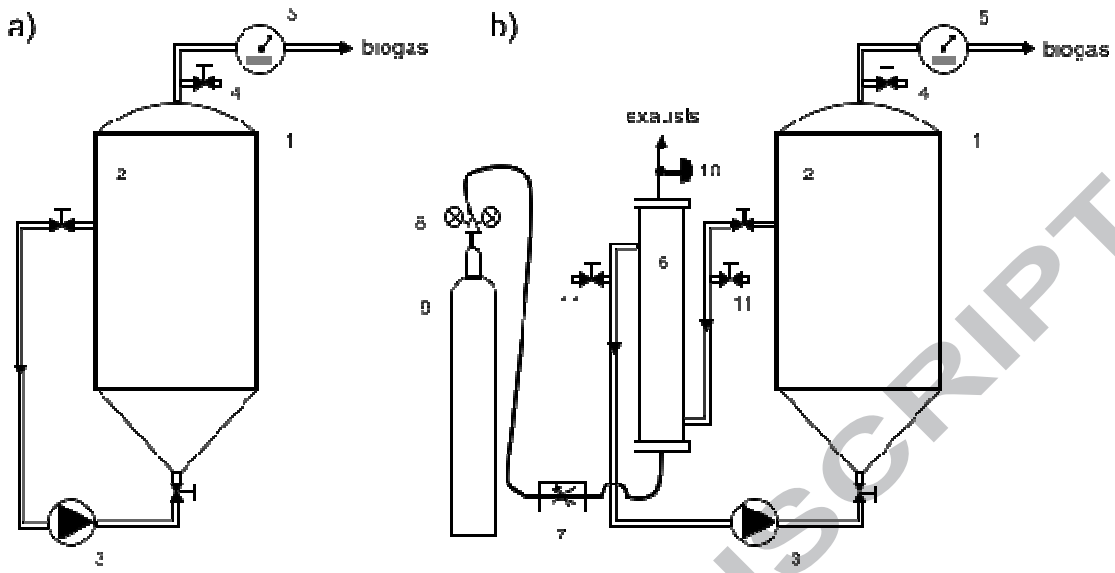

Figure 1. Scheme of the experimental rig. (a) Control reactor and (b) Test reactor configuration during CO_2 injection. (1) Anaerobic reactor, (2) heating jacket, (3) peristaltic pump, (4) biogas sample point, (5) biogas meter, (6) bubble column, (7) mass flow controller, (8) gas pressure regulator, (9) CO_2 cylinder, (10) CH_4-CO_2 analyser, (11) digestate sampling point.

ACCEPTED MANUSCRIPT

ACCEPTED MANUSCRIPT

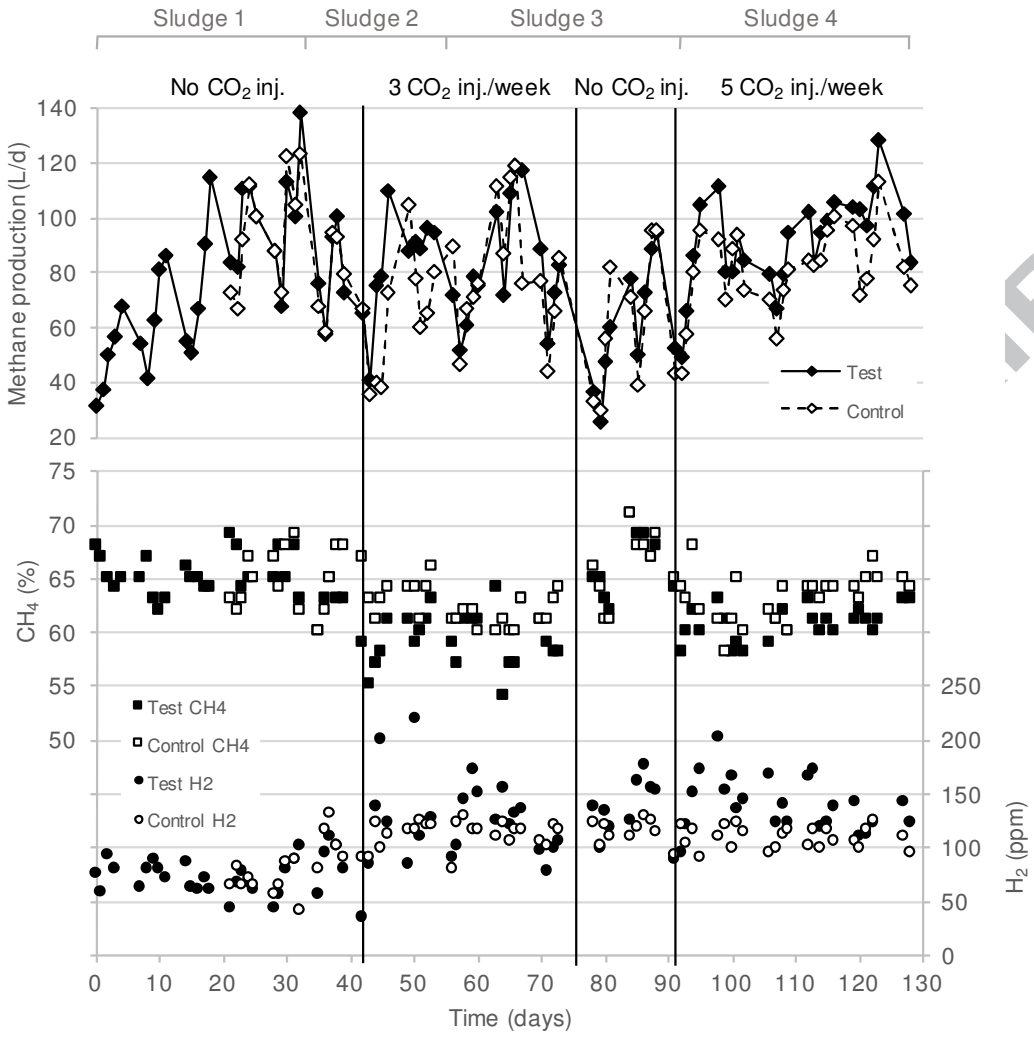

Figure 2. Methane (CH_4) production, CH_4 and hydrogen (H_2) concentration in Test and Control reactors during the experimental period. Black vertical lines divide the phases of the experimental period between: no CO_2 injections phase (No CO_2 inj.), phase of moderate CO_2 enrichment at 3 injections per week with a bubble column (3 CO_2 inj./week) and phase of intense CO_2 enrichment at 5 injections per week with a packed column (5 CO_2 inj./week). Top grey line identifies when different samples of sludge were used.

ACCEPTED MANUSCRIPT

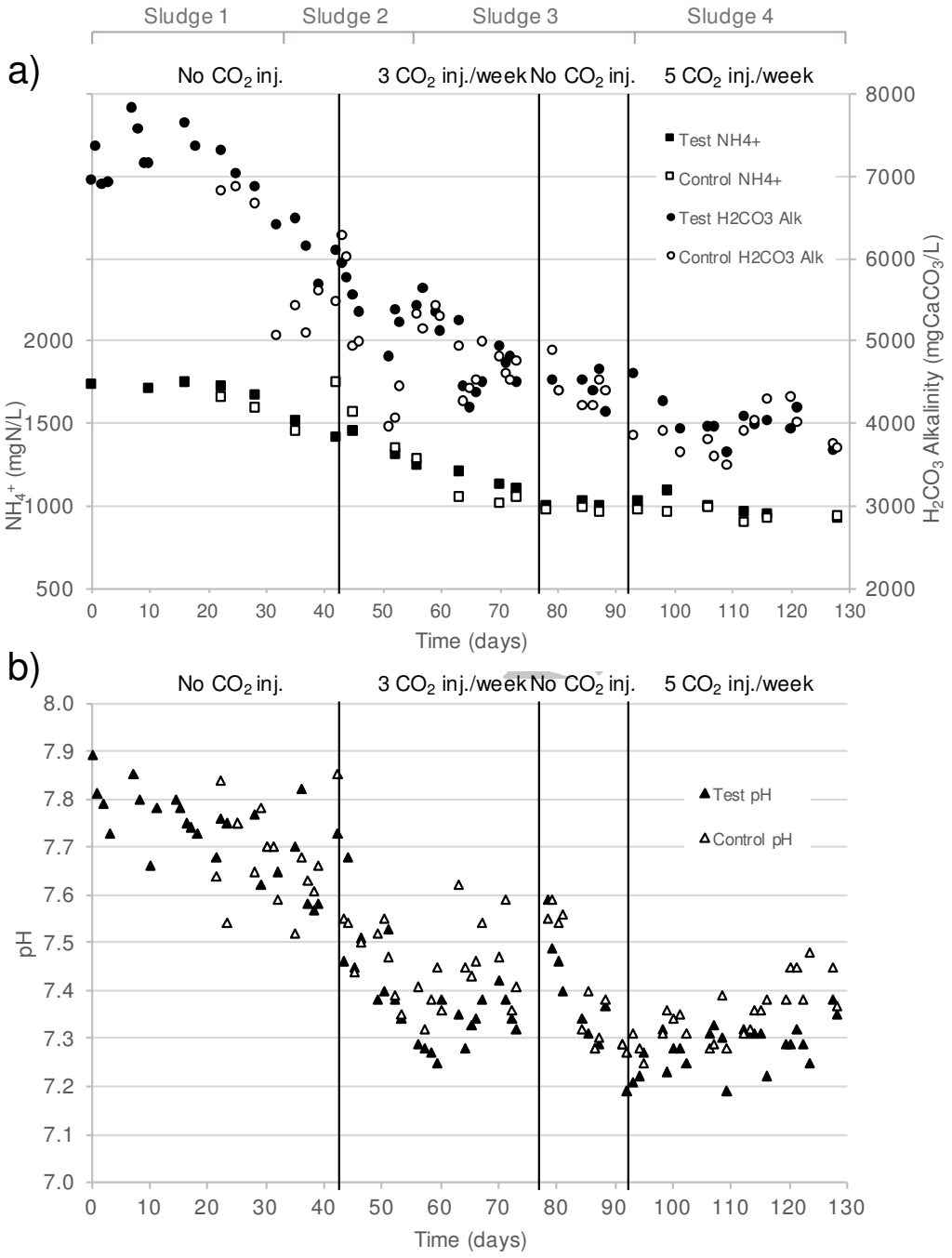

Figure 3. Ammonium nitrogen (NH_4^+), H_2CO_3 Alkalinity concentrations (a) and pH (b) for Test and Control reactors during the different phases of the experimental period. Vertical lines divide the phases of the experimental period. Top grey line identifies when different samples of sludge were used.

ACCEPTED MANUSCRIPT

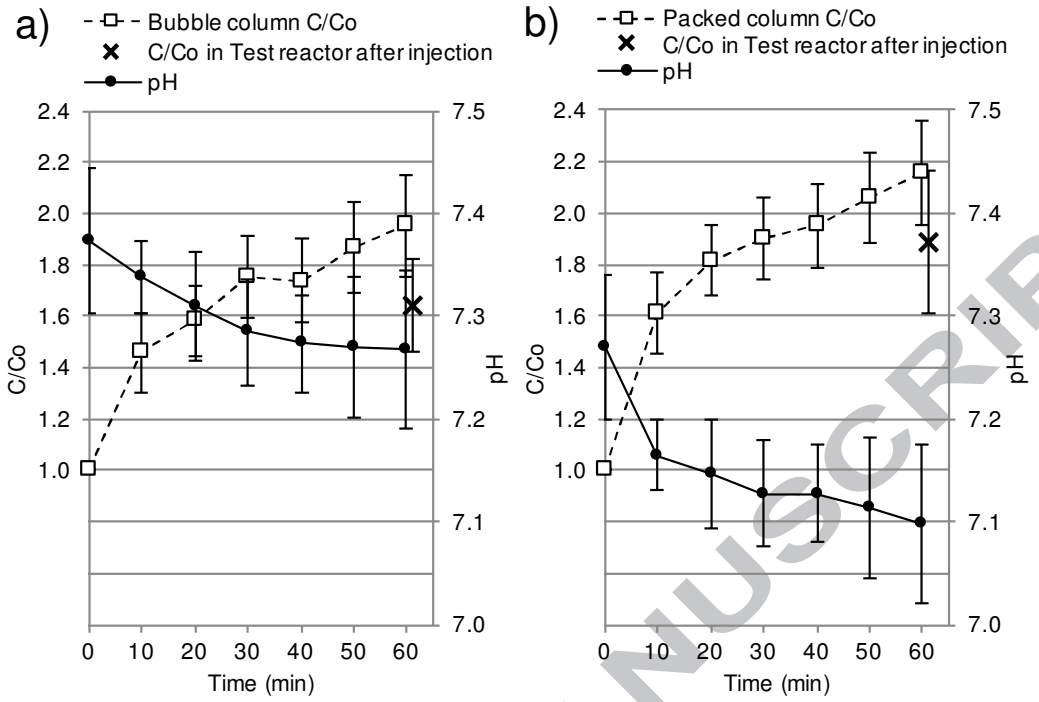

Figure 4. Evolution of the parameter C/C_0 representing the ratio between the initial CO_2 concentration in digestate (C_0) and the concentration on the effluent of the CO_2 injection column (C). Evolution of pH in the effluent of the CO_2 injection column. The C/C_0 achieved in the Test reactor at the end of the injection is marked as "X". Graph a) is for the use of a bubble column, graph b) is for the use of a packed column.

ACCEPTED MANUSCRIPT

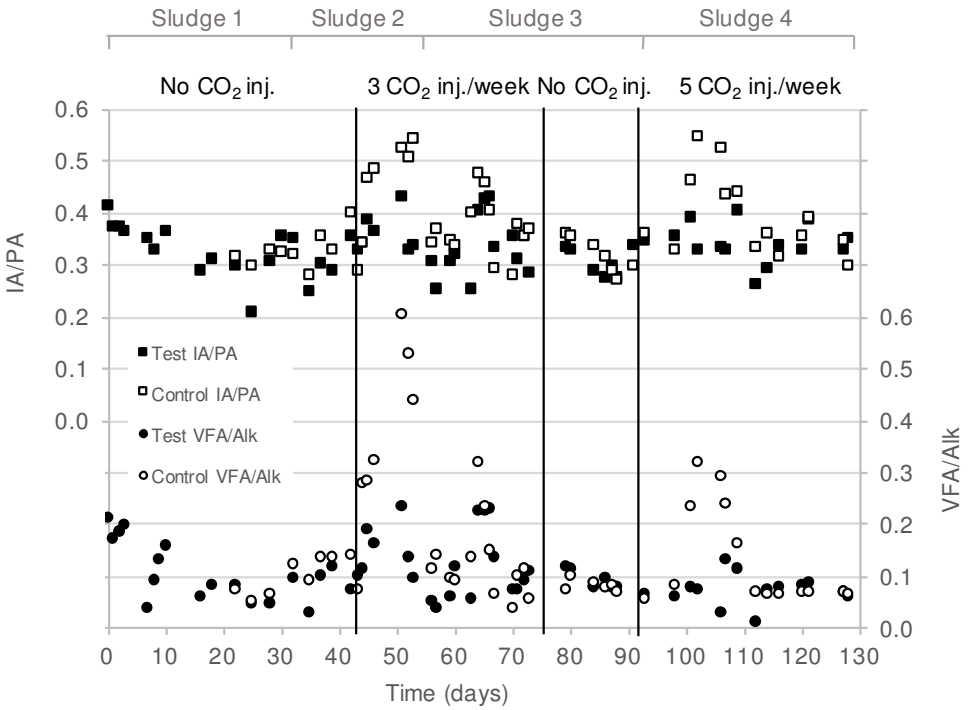

Figure 5. Intermediate to Partial Alkalinity (IA/PA) ratio and volatile fatty acids to H_2CO_3 Alkalinity (VFA/Alk) ratio for Test and Control reactors during the different phases of the experimental period. Vertical lines divide the phases of the experimental period. Top grey line identifies when different samples of sludge were used.

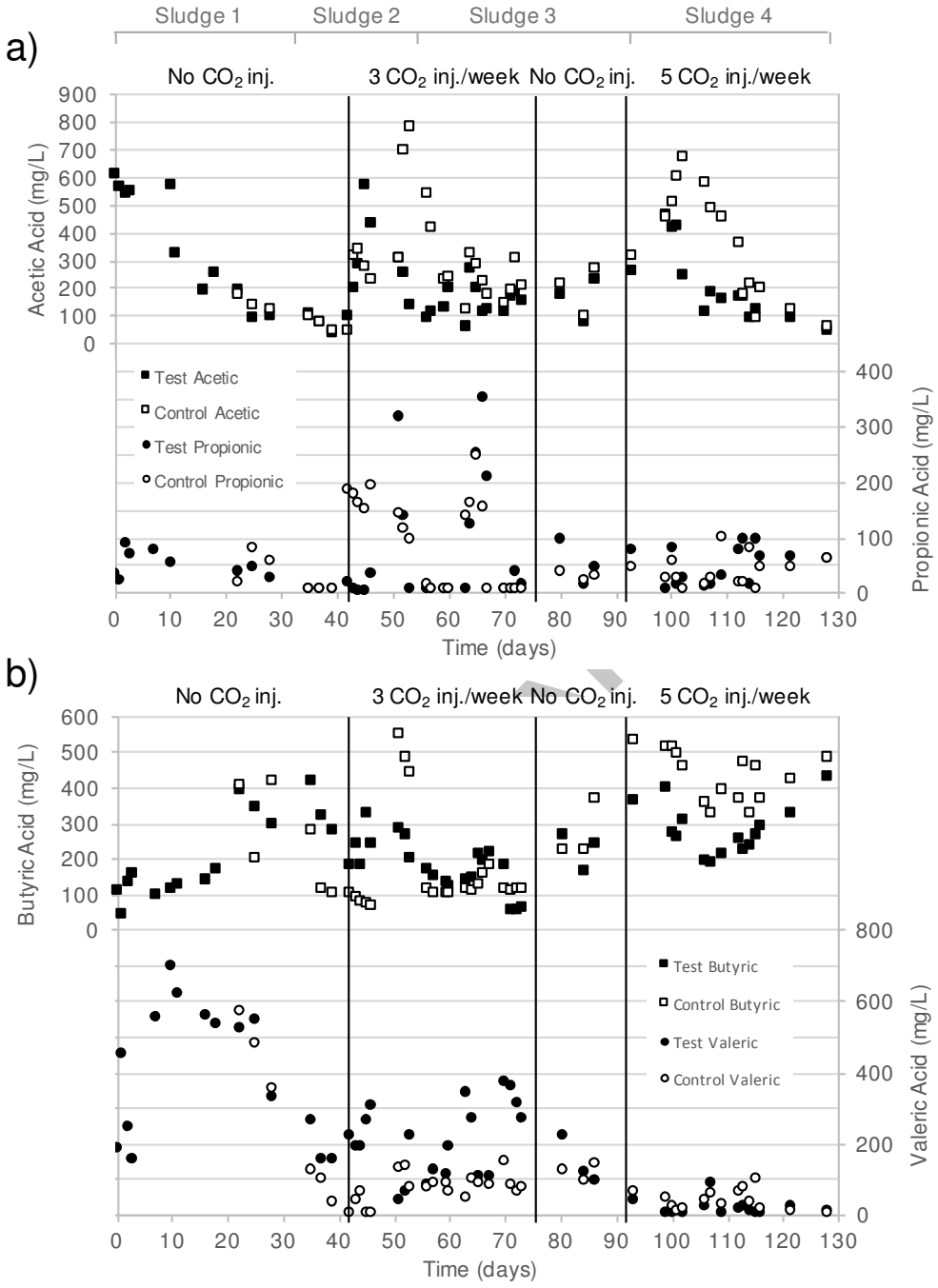

Figure 6. Acetic and propionic acid concentrations (a) and butyric and valeric acid concentrations (b) for Test and Control reactors during the different phases of the experimental period. Vertical lines divide the phases of the experimental period. Top grey line identifies when different samples of sludge were used.

ACCEPTED MANUSCRIPT

HIGHLIGHTS

- CO_2 enrichment was tested on sewage sludge anaerobic digestion at pilot scale.
- CO_2 enrichment enhanced CH_4 production under moderate and intense injections.
- CO_2 injection had no negative effects on anaerobic digestion process stability.
- Benefits of CO_2 enrichment were proved without exogenous H_2 addition.

ACCEPTED MANUSCRIPT

https://dspace.org/

Performance and stability of sewage sludge digestion under CO2 enrichment: a pilot study

Alibardi, Luca; Green, Kevin; Favaro, Lorenzo; Vale, Peter C. J. ; Soares, Ana ...

2017-08-16
Elsevier
http://dx.doi.org/10.1016/j.biortech.2017.08.071
Alibardi L, Green K, Favaro L, et al., (2017) Performance and stability of sewage sludge digestion under CO2 enrichment: a pilot study. Bioresource Technology, Volume 245, Part A, December 2017, pp. 581-589

Downloaded from a DSpace repository. https://dspace.org/